

# Micro-mixing performance of mesoscale oscillatory baffled reactors

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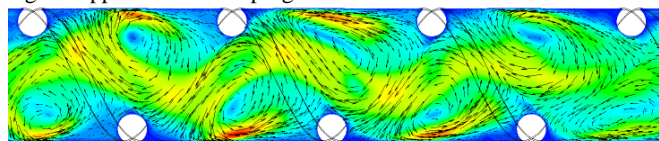
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## 1. Introduction

The mesoscale oscillatory baffled reactor (meso-OBR) is a novel technology for reaction engineering and screening applications. The meso-OBR typically consists of a 5 mm diameter tube fitted with one of a variety of “plug-and-play” tube insert geometries in which oscillatory motion of the liquid is superimposed on to the net flow. The tube inserts act as flow-constricting devices that increase shear forces in the flow, disrupting the boundary layer at the wall, whilst oscillatory motion in combination with these inserts results in the formation of vortices upon each flow reversal (Figure 1). Consequently, the meso-OBR behaves as many well-mixed tanks-in-series, providing a good approximation of plug flow.



**Figure 1.** Example of vortex formation around a helical coil tube insert design in an oscillatory flow

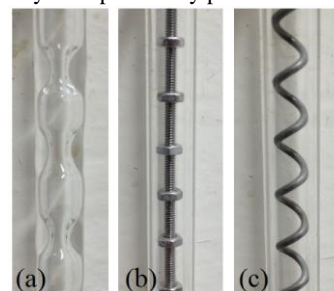
Superposition of the oscillatory motion with the net flow of liquid results in a decoupling of the mixing intensity from the net flow rate. Consequently, plug flow can be achieved at laminar net flow rates. This allows them to operate at flow rates on the order of mL/h, making them competitive with the widely adopted microreactor. Numerous examples of using the meso-OBR as a flow chemistry screening platform have been demonstrated [1].

The plug flow behavior of these reactors has been widely studied [2-4]. However, it has been longed assumed that this good plug flow equates with good mixing, which is not necessarily the case. Therefore, this study aims to address the limited understanding of mixing in these meso-OBRs, with the current focus being the micro-mixing behavior. The objective of the present work is to therefore perform the Villermaux-Dushman (iodide/iodate) reaction in various meso-OBR designs subject to a wide range of net flow rates and oscillation intensities, and compare the resulting micro-mixing times with the plug flow behavior previously published [2-4].

## 2. Methodology

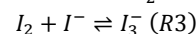
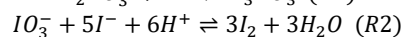
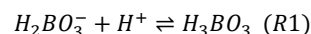
Three tube insert designs were considered in the present study: integral, central and helical (shown in Figure 2). Each insert type was tested in a glass tube with 5 mm i.d. and 350 mm length. The integral baffles were approximately equally spaced every 7.5 mm and had an orifice diameter of 2.5 mm. The central baffles were also spaced every 7.5 mm, and fabricated by threading 4 mm

stainless steel nuts onto a 1.2 mm diameter stainless steel studding. Finally, the helical coil was created from a 1 mm diameter stainless steel wire coiled with a pitch of 7.5 mm. These designs maintained geometric similarity with previously published studies [2-4].



**Figure 2.** Meso-OBR tube insert designs / (a) integral, (b) central and (c) helical

The Villermaux-Dushman reaction scheme consists of two competing reactions. The redox reaction (R2) is fast, and occurs on the same time scale as mixing, provided the concentrations of the chemicals are appropriately selected [5]. The neutralization reaction (R1) can be considered instantaneous in comparison to R2. Thus, by contacting a solution of iodide ( $I^-$ ) and iodate ( $IO_3^-$ ) in a  $H_2BO_3/H_3BO_3$  buffer with an acid solution, knowledge about the mixing state can be obtained. Under ideal mixing, the acid is only consumed by R1 ( $R1 \gg R2$ ), meaning there are no residual protons to produce  $I_2$  in R2. However, under poor mixing conditions, the dissipation time of the acid is larger than the reaction time of R1; local over-concentrations of the acid occur once  $H_2BO_3^-$  has been locally protonated, allowing for the formation of  $I_2$  in the second reaction. The  $I_2$  then reacts with  $I^-$  to produce  $I_3^-$  (R3) that can be detected via UV-Vis spectrometry.



In this study, the Villermaux-Dushman reaction was applied using the approach recommended by Commenge and Falk (2011) [5]. First the magnitude of the mixing times in the meso-OBR were predicted using equation 1, derived by correlating the mixing times against the energy dissipation rate using literature data [5]. For the meso-OBR, the quasi-steady state model (equation 2) was used to estimate the dissipation rate ( $\epsilon$ ) at each oscillation intensity [6]. Here,  $N$  is the number of equivalent tanks-in-series,  $f$  is the oscillation frequency,  $x_0$  is the oscillation amplitude,  $S$  is the fractional open area of the tube insert,  $C_0$  is the orifice discharge coefficient and  $Z$  is the tube length.

$$t_m = 0.15\epsilon^{-0.45} \quad (1)$$

$$\varepsilon = \frac{2N(2\pi f x_o)^2(1/S^2 - 1)}{3\pi C_o^2 Z} \quad (2)$$

Then, based on this expected mixing time, the most suitable concentration set was selected so that the expected optical density measured by the UV-Vis spectrometer would be around 1 for a 10 mm path length cuvette [5]. Reactions were then performed at the desired flow rate and oscillation intensity, with a 1 mL sample of the reaction mixture collected at the outlet after the reactor reached steady state (at least two residence times). The optical density of this sample at a wavelength of 353 nm was finally measured, which was subsequently used to calculate the mixing time.

All reagents were used as supplied from Sigma Aldrich, and were prepared at the desired concentrations in deionized water. The iodide/iodate solution was prepared using potassium iodide, potassium iodate, boric acid and sodium hydroxide. The acid solution was prepared using sulfuric acid, meaning the error in the mixing times was likely to be approximately a factor of 2 (i.e. mixing times that differed by less than a factor of two are likely to be within the error) [5]. The solutions were run at the same net flow rate to ensure the correct stoichiometry, and it was confirmed by taking multiple measurements of collected samples that the reaction was complete before the sampling point.

Reagent net flow rates and fluid oscillation were achieved using C3000 series syringe pumps as described previously [2-4]. In this study, net flow Reynolds numbers ( $Re_n$ ) of 5–40 and oscillatory Reynolds numbers ( $Re_o$ ) of 0–1000 were considered. These are shown in equations 3 and 4 respectively, where  $\rho$  is the liquid density,  $v$  is the superficial net flow velocity,  $D$  is the tube diameter,  $\mu$  is the liquid viscosity,  $f$  is the oscillation frequency and  $x_o$  is the oscillation amplitude.

$$Re_n = \frac{\rho v D}{\mu} \quad (3)$$

$$Re_o = \frac{2\pi f x_o \rho D}{\mu} \quad (4)$$

### 3. Results to Date

Figure 3 shows the micro-mixing times produced at different  $Re_n$  when increasing the oscillation intensity ( $Re_o$ ) using the helical insert design (Figure 1c). It can be seen that higher mixing intensities expectedly produce smaller mixing times. As the oscillation intensity around the helical coils increases, the radial flow as well as swirling intensity increases [7]. This likely leads to much shorter diffusion path lengths for mixing through the continual refolding of the fluid streamlines in the oscillatory flow. Overall, micro-mixing times of 0.35–20.89 s were produced.

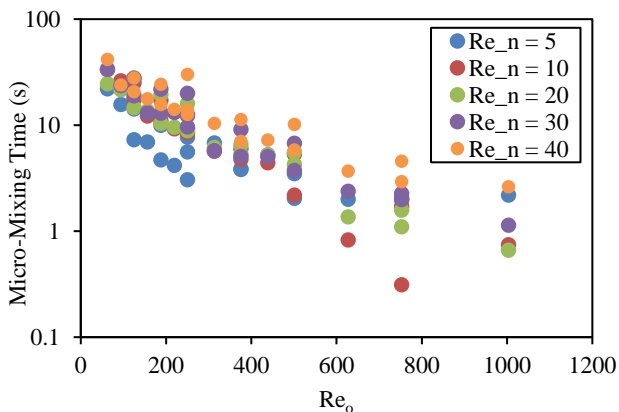


Figure 3. Micro-mixing times vs mixing intensity ( $Re_o$ )

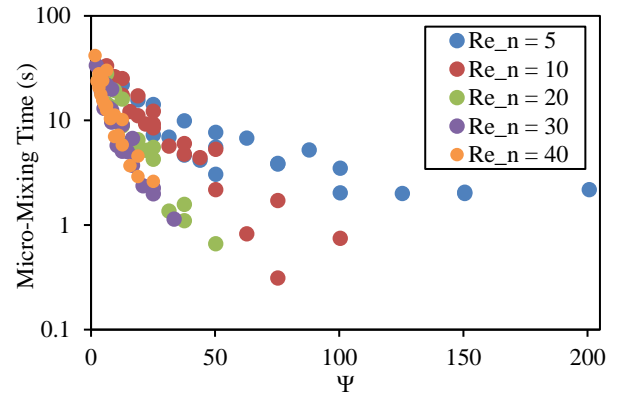


Figure 4. Micro-mixing times plotted against velocity ratio

In Figure 4, the same data as Figure 3 has been plotted against the velocity ratio,  $\Psi$ , defined as  $Re_o/Re_n$ . Here, it can be seen that for  $Re_n \geq 20$  the micro-mixing trends all align along a single design curve. However, the trends for  $Re_n = 5$  and 10 are shallower, implying that the increase in oscillation intensity is less effective at reducing the mixing times at these lower flow rates.

### 4. Ongoing and Future Work

Experiments are currently ongoing. The immediate focus is repeating some of the conditions reported in Figures 3 and 4 in order to quantify the magnitude of the experiment error. Following this, similar micro-mixing times will be determined for the integral and central insert designs. These will then be compared with the plug flow results previously reported in order to establish whether the previous interchange of the term “plug flow” with “mixing” in the literature is valid. The roles of diffusion and convection at these mesoscales will also be attempted to be elucidated.

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